

VELOCITY OF SOUND OF SATURATED R 152a

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ABSTRACT

The paper features the mathematical model of computing velocity of sound in the liquid and gas domain for R 152a with the help of classical thermodynamics and statistical thermodynamics. To calculate the thermodynamic properties of real fluid the Johnson-Zollweg-Gubbins model based on the modified Benedict-Webb-Rubin equation of state, Chunxi-Yigui-Jiufang equation of state based on simple perturbation theory, complex Tang-Tong-Lu model based on solution the Ornstein-Zernike equation with help of perturbation theory and Mecke-Müller-Winkelman-Fischer on the base of generalized VDW equation of state, was applied. To calculate the thermodynamic properties of real fluid with the help of classical thermodynamics we used Van Der Waals, Peng-Robinson, Benedict-Webb-Rubin, Benedict-Webb-Rubin-Starling-Nishiumi, Jacobsen-Stewart and Tilner-Roth equation of state. We developed the mathematical model for the calculation of all equilibrium thermodynamical functions of state for pure refrigerants and their mixtures. The analytical results obtained by classical and statistical thermodynamics are compared with the experimental data and they show relatively good agreement.

Introduction

The calculation of thermodynamic properties of state of refrigerants is exceptionally important in cooling system processes design. In engineering practice the refrigeration processes calculation is most often carried out by means of charts, diagrams or pre-designed computer programs. Looking for thermodynamic properties using diagrams and charts is extremely time-consuming. With computer programs, on the other hand, the time required for data collection is substantially shortened, thus enabling us to calculate some of the most frequently employed refrigerants, although computer programs allow only the calculation of certain thermodynamic properties, such as entropy, pressure and enthalpy of mixture. In practice we also often use equations of state, obtained through classical thermodynamics. Classical thermodynamics calculates the thermodynamic properties of state on the basis of macroscopic observation of substances. The equations obtained by means of classical thermodynamics are empirical and apply only in the region under observation. The main drawback of classical thermodynamics is that it lacks the insight into the substance of microstructure. Contrary to classical thermodynamics, statistical thermodynamic calculates the thermodynamic properties of state on the basis of intermolecular and intramolecular interactions between particles in the same system of molecules. It deals with the systems composed of a very large number of particles.

Refrigerants

Refrigerants are working fluids transferring heat from lower temperature level to a higher temperature level. In accordance with the second main law of thermodynamics this implies the use of work. Refrigerants must meet certain requirements. In particular it is important that they change the state of aggregation and this must be accompanied by adequate temperature and pressure changes. Water belongs to the first cooling mediums and is still used in vapour systems. In 1876, ammonia appeared as the first commercial and technically important refrigerant, followed by SO_2 , CO_2 (R744)... When halogenated refrigerants (freons) began to be widely used in 1928 this provided the impetus in the research of refrigerants. Halogenated refrigerants belong to halogens of lower aliphatic cyclically saturated hydrocarbons. Freons are inflammable, at normal conditions they are mostly gases, the boiling point is between -100°C and 0°C , when compressed they easily liquefy, they are not explosive if mixed with air, are non toxic, are odourless and cause no irritation, do not affect foodstuffs, are chemically stable and have favourable thermodynamic properties. Thus, used as refrigerants they have an absolute advantage with the only exception being ammonia. Known are also azeotropic mixtures, near azeotropic and zeotropic mixtures. As to the evaporation and condensation azeotropic mixtures behave similarly as pure substances and have dramatically extended the limits of operation. Today, the use of refrigeration mixtures is rapidly increasing. In Europe, the R502 is the third most widely used refrigerant. In general, any substance can be used as a refrigerant. The differences between various refrigerants exist due to various physical, chemical and physiological properties.

For several decades chlorine-fluorine-hydrocarbons were considered harmless refrigerants. In 1968, however, the first serious ozone layer damage was observed. According to the meteorologists' findings it resulted from a dramatic increase in the emission of chlorine- fluorine- hydrocarbons. In the recent period, the greenhouse effect has also become substantially more evident.

1,1 Difluoroethane (HFC-152a) is an alternative refrigerant which is suitable as a substitute for CFC-12. Due to its flammability, this fluid has not seen widespread use in the refrigeration and air-conditioning industry. However, its zero ODP (ozone depletion potential) and very low GWP (greenhouse warming potential) make it an interesting ozone friendly refrigerant for applications where its hazards can be kept under control.

Furthermore, it is used as a component of mixtures to enhance cycle performance and to lower GWP of the total charge of a refrigeration cycle.

Classical Thermodynamics

In most cases the thermodynamic tables or diagrams or different empirical functions obtained from measurement are used (classical thermodynamics). Today, there are numerous equations of state (EOS) reported in the literature for describing the behaviours of fluid [1]: (Van der Waals EOS (VDW), Peng-Robinson (PR), Redlich-Kwong EOS (RK), Soave EOS..). However, these equations have exhibited some noticeable defects, such as poor agreement with experimental data at moderate densities. On the other side, we can use the complex equations of state with many constants (Benedict-Webb-Rubin [1] (BWR) EOS, Benedict-Webb-Rubin-Starling-Nishiumi [1,2,3] EOS (BWRSN), Tilner-Roth (TR) [4] EOS, Jacobsen-Stewart EOS [5,6] (JS)...). These equations are more complicated but they have no insight into the microstructure of matter and poor agreement with experimental data outside interpolation limits. The calculation of thermodynamical functions of state with help of classical thermodynamics is well-known matter and is not described in this paper. Table 1 shows only the fundamental characteristics of various EOS obtained by classical thermodynamics.

For the caloric equation of state of ideal gases (ig) we used the following formulation:

$$C_v^{ig} = R_m \left(a_1 + a_2 T + a_3 T^2 + a_4 / T^2 - 1 \right). \quad a_1, a_2, a_3, a_4 \dots \text{const.} \quad (1)$$

For the real calculation of specific heats at a constant volume C_v of one-component systems we used the known expression of classical thermodynamics.[1]:

$$C_v = C_v^{ig} + \left[T \int_{V_\infty}^V \left(\frac{\partial^2 p}{\partial T^2} \right) \cdot dV \right]_{T, \psi} \quad (2)$$

The expression V_∞ denotes the volume of a kilogram molecule of substance, where the gas is ideal. To calculate thermodynamic properties of state in mixtures we employed the method of calculation of thermodynamic properties of state using the fugacity coefficients. The calculation procedure was carried out in the following manner:

1 Calculation of fugacity coefficients:

$$\ln \phi_i = \int_0^p \left(\frac{\bar{V}_i}{R_m T} - \frac{1}{p} \right) dp \quad (3)$$

2. Calculation of chemical potentials:

$$\mu_i = \mu_{0i}(p^+, T) + R_m T \cdot \ln \left(\frac{p_i}{p^+} \right) + R_m T \cdot \ln(\phi_i) \quad (4)$$

In Eq. (4) p^+ is the reference pressure, p_i is the partial pressure, $\mu_{0i}(p^+, T)$ is the chemical potential of the pure constituent i .

3. Calculation of partial properties of state:

4. Calculation of thermodynamic properties of state for mixtures:

Statistical thermodynamics

To calculate thermodynamic functions of state we applied the canonical partition [7]. Utilising the semi-classical formulation for the purpose of the canonical ensemble for the N indistinguishable molecules the partition function Z can be expressed as follows [7]:

$$Z = \frac{1}{N! h^{Nf}} \int \dots \int \exp \left(-\frac{H}{kT} \right) \cdot d\vec{r}_1 d\vec{r}_2 \dots d\vec{r}_N d\vec{p}_1 d\vec{p}_2 \dots d\vec{p}_N, \quad (1)$$

where f stands for the number of degrees of freedom of individual molecule, H designates the Hamiltonian molecule system, vectors $\vec{r}_1, \vec{r}_2 \dots \vec{r}_N \dots$ describe the positions of N molecules and $\vec{p}_1, \vec{p}_2 \dots \vec{p}_N$ momenta, k is Boltzmann's constant and h is Planck's constant. The canonical ensemble of partition function for the system of N molecules can be expressed like this:

$$Z = Z_0 Z_{\text{trans}} Z_{\text{vib}} Z_{\text{rot}} Z_{\text{ir}} Z_{\text{el}} Z_{\text{nuc}} Z_{\text{conf}}. \quad (2)$$

Thus the partition function Z is a product of terms of the ground state (0), the translation (trans), the vibration (vib), the rotation (rot), the internal rotation (ir), the influence of electrons excitation (el), the influence of nuclei excitation (nuc) and the influence of the intermolecular potential energy (conf).

Utilising the canonical theory for computing the thermodynamic functions of the state can be put as follows:

$$\begin{aligned}
 \text{Pressure } p &= kT \left(\frac{\partial \ln Z}{\partial V} \right)_T . \\
 \text{Freeenergy } A &= -kT \cdot \ln Z . \\
 \text{Entropy } S &= k \left[\ln Z + T \left(\frac{\partial \ln Z}{\partial T} \right)_V \right] . \\
 \text{Enthalpy } H &= kT \left[T \left(\frac{\partial \ln Z}{\partial T} \right)_V + V \left(\frac{\partial \ln Z}{\partial V} \right)_T \right] ,
 \end{aligned} \tag{3}$$

where T is temperature and V is volume of molecular system. The computation of the individual terms of the partition function and their derivatives except of the configurational integral is dealt with in the works of Lucas [7] ; Gray and Gubbins [8] and McClelland [9].

The various derivatives and expressions of the fundamental equations (3) have an important physical significance. In this paper we focused on the calculation of velocity of sound. The term velocity of sound refers to the velocity of the mechanical longitudinal pressure waves propagation through a medium. It is very important parameter in the study of compressible fluids flows and in some applications of measurement (acoustic resonance level gauge).

The propagation of sonic waves for real fluids is almost in all cases nearly isentropic. Therefore, we can calculate the isentropic speed of sound for the real fluid c_0 :

$$c_0 = \sqrt{-V^2 \left(\frac{\partial p}{\partial v} \right)_{s, \psi} \frac{1}{M}} = \sqrt{-\frac{V^2}{M} \frac{\frac{c_p}{T} \left(\frac{\partial T}{\partial V} \right)_{p, \psi}}{\left(\frac{\partial V}{\partial T} \right)_{p, \psi} - \frac{C_p}{T} \left(\frac{\partial T}{\partial p} \right)_{v, \psi}}}, \quad (4)$$

where T is temperature, p is pressure, S is entropy, M is molecular mass, C_p is the molar heat capacity at constant pressure and ψ is molar concentration.

Configurational integral

a) Johnson-Zollweg-Gubbins (JZG) model [10]

For a real fluid the Johnson-Zollweg-Gubbins model based on molecular dynamics and Monte Carlo simulations with the Lennard-Jones intermolecular potential is possible to use. The modified BWR EOS contains 32 linear parameters (x_i) and one non-linear parameter (γ) [10]. On this basis we can express configurational free energy A_{conf} :

$$A_{\text{conf}}^* = \sum_{i=1}^8 \frac{a_i \rho^{*i}}{i} + \sum_{i=1}^6 b_i G_i, \quad (5)$$

where the coefficients a_i , b_i and G_i are presented in Reference [10]. The coefficients a_i and b_i are functions of reduced temperature T^* only, the coefficients G_i are function of the reduced density ρ^* and nonlinear adjustable parameter γ .

$$\rho^* = \frac{N\sigma^3}{V}, \quad T^* = \frac{kT}{\varepsilon}, \quad A_{\text{conf}}^* = \frac{A_{\text{conf}}}{N\varepsilon}, \quad F = \exp(-\gamma \rho^{*2}), \quad \gamma=3. \quad (6)$$

where A_{conf} is reduced configurational free energy, σ and ε are Lennard-Jones parameters. Equation (5) accurately correlates the thermophysical properties from the triple point to about 4 to 5 times the critical temperature. With help of configurational free energy we can calculate all configurational thermodynamic properties. Expressions

for calculation of configurational entropy and internal energy are shown in literature [10]. We carried out all other expressions for calculation of thermophysical properties.

b) Revisited Cotterman model (CYJ) [11,12]

Revisited Cotterman EOS is based on the hard sphere perturbation theory. The average relative deviation for pressure and internal energy in comparison with Monte-Carlo simulations are 2.17% and 2.62% respectively for 368 data points [11,12]. The configurational free energy is given by:

$$A_{\text{conf}} = A^{\text{hs}} + A^{\text{pert}}, \quad (7)$$

$$\frac{A^{\text{hs}}}{R_m T} = \frac{4\eta - 3\eta^2}{(1 - \eta)^2}, \quad A^{\text{pert}} = \frac{A^{(1)}}{T^*} + \frac{A^{(2)}}{T^*}, \quad (8)$$

$$\frac{A^{(1)}}{R_m T} = \sum_{m=1}^4 A_{1m} \left(\frac{\eta}{\tau} \right)^m, \quad \frac{A^{(2)}}{R_m T} = \sum_{m=1}^4 A_{2m} \left(\frac{\eta}{\tau} \right)^m, \quad (9)$$

$$\tau = 0.7405, \quad \eta = \frac{\pi \rho D^3}{6}, \quad (10)$$

where η is packing factor, D is hard-sphere diameter. With help of configurational free energy we can calculate all configurational thermodynamic properties. Expressions for calculation of configurational entropy and internal energy are shown in literature [12]. We carried out all other expressions for calculation of thermophysical properties.

c) Tang-Tong-Lu model

Tang-Tong-Lu model uses as the intermolecular potential a new two-Yukawa function. This function is found to mimic very closely the Lennard-Jones potential. Tang-Tong-Lu analytical model calculates thermodynamic functions of state on the base of salvation of

the Ornstein-Zernike equation with help of perturbation theory. Comparisons with the computer simulation data indicate that the expressions developed yield better results (pressure, internal energy, free energy) than theory of Weeks et al. [13], Barker and Henderson [14]. Results of pressure, internal energy and free energy obtained by TTL model are comparable with the most recent modified Benedict-Webb-Rubin EOS. The configurational free energy is given by [15]:

$$\frac{A_{\text{conf}}}{NkT} = a_0 + a_1 + a_2, \quad (11)$$

where a_0 represents the free energy of the hard sphere fluid, a_1 and a_2 are perturbed first and second order parts.

d) Mecke et al. [16,17] (MMWF) model

The presented model is obtained on the base of generalized van der Waals EOS, Setzmann-Wagner optimization procedure and hybrid Barker-Henderson perturbation theory. On this basis we can express A_{conf} :

$$\frac{A_{\text{conf}}}{NkT} = \frac{4\zeta - 3\zeta^2}{(1 - \zeta)^2} + \sum_{i=1}^{32} c_i \left(\frac{T^*}{T_c^*} \right)^{m_i} \left(\frac{\rho^*}{\rho_c^*} \right)^{n_i} \exp \left[p_i \left(\frac{\rho^*}{\rho_c^*} \right)^{q_i} \right]. \quad (12)$$

The presented equation covers the whole fluid region up to highest densities in the temperature range $0.7 < T^* < 10$ with high accuracy. The MMWF EOS contains 160 constants. In Eq. (3) ζ represents the packing factor, T_c^* and ρ_c^* are critical reduced temperature and density.

Results and comparison with experimental data

The constants necessary for the computation such as the characteristic rotation-, vibration-, electronic etc. temperatures are obtained from experimental data [18,19,20]. The inertia moments are obtained analytically by applying the knowledge of the atomic structure of the molecule. Constants for Lennard-Jones potential are obtained from the literature [8,21].

Table 2 represents the deviation of the results for the velocity of sound on the saturated curve between the analytical computation (ST- statistical theory with help of classical virial expansion [22]), JZG-Johnson-Zollweg-Gubbins model, TTL-Tang-Tong-Lu model, CYJ-Chunxi-Yigui-Jiufang model, MMWF-Mecke et al. model), models obtained by classical thermodynamics (Table 1) and the experimental values (Exp) [23].

Table 3 shows the deviation of the results on the boiling curve for R 152a between the analytical computation and experimental values [24,25]. The experimental data are obtained by dynamic light scattering techniques [26].

The results for all models obtained by statistical thermodynamics show very good agreement. The computed vapour velocity of sound conform well for all models, obtained by statistical thermodynamics, with the measured vapour pressure and velocity of sound. Somewhat larger deviations can be found in the region of critical conditions due to the large influence of fluctuation theory [27] and singular behaviour of some thermodynamic properties in the near-critical condition. Larger deviations can however be found in the region of real liquid due to the large influence of the attraction forces, since the Lennard-Jones potential is only an approximation of the actual real intermolecular potential. The best results in the liquid-gas domain are obtained with the classical JS model. Surprisingly good results are obtained by simple CYJ model. The

comparison between computed analytical results obtained by the statistical thermodynamics and measured one shows slightly less good agreement of results.

Conclusion And Summary

The paper presents the mathematical model for computation of thermodynamically functions of the state in the liquid and gaseous region. For the real fluid the Johnson-Zollweg-Gubbins model based on molecular dynamic and Monte Carlo simulations and modified BWR EOS, Tang-Tong-Lu complex perturbation model and Chunxi-Yigui-Jiufang model and Mecke et al. Model, was applied. We developed the expressions for the calculation the velocity of sound.

The analytical results are compared with the experimental results and analytical calculation obtained by classical thermodynamics and show relatively good agreement. In the region of real gases the results are an equally good match.

In comparison with experimental data, somewhat larger deviations can however be found in the region of real liquid

List os symbols

A free energy

A^* reduced free energy

BWR Benedict-Webb-Rubin

BWRSN Benedict-Webb-Rubin-Starling-Nishiumi

c_0 velocity of sound

C_p heat capacity at constant pressure per mole

C_v heat capacity at constant volume

CYJ	Chunxi-Yigui-Jiufang model
D	hard sphere diameter
E	potential energy
EOS	equation of state
Exp	experiment
f	number of degrees of freedom
G	free enthalpy
H	enthalpy
h, \hbar	Planck constant
JS	Jacobsen-Stewart
JZG	Johnson-Zollweg-Gubbins
k	Boltzmann constant
M	molecular mass
N	number of molecules in system
p	pressure, momentum
PR	Peng-Robinson
R_m	universal gas constant
RK	Redlich-Kwong
S	entropy
ST	statistical virial theory
T	temperature
T^*	reduced temperature
TR	Tilner-Roth
TTL	Tang-Tong-Lu model

V	volume
VDW	Van der Waals
Z	partition function
ε	Lennard-Jones parameter
η	packing factor
μ	chemical potential
ψ_i	molar fraction
ρ	density
ρ^*	reduced density
σ	Lennard-Jones parameter

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TABLES

Table1. Fundamental characteristics of various EOS obtained on the base of classical thermodynamics.

EOS	Number of used constants	Influence of polarity	Region
VDW	2	NO	Real gas, mixtures
RK	2	NO	Real gas
PR	3	YES	Real gas, Phase transition
BWR	8	NO	Real gas
BWRSN	16	YES	Real gas, real liquid
TR	62	YES	Real gas, real liquid
JS	40-150	YES	Real gas, Real liquid

Table 2: Velocity of sound on the evaporation curve

T K	Exp.	JS	BWRSN	ST	JZG	TTL	CYJ	MMWF	CT	TR
203	169.9	172.86	173.4	166.2	166.5	167.7	166.1	168.1	166.3	172.6
213	174.6	176.8	177.7	170.1	170.4	170.4	170.4	169.8	170.0	175.9
223	178.4	179.1	180.6	173	173.8	173.8	173.8	173.1	173.4	178.8
233	181.3	181.6	183.8	176	176.0	176.2	176.0	176.1	176.7	181.3
243	183.6	183.7	186.6	178.7	178.7	179.0	178.7	178.9	179.7	183.4
253	185.3	185.35	189	181	181.0	181.4	180.9	181.2	182.4	185.1
263	186.4	186.42	191	182.9	182.8	183.4	182.9	183.2	183.3	186.2
273	187	186.9	192.4	184.2	184.1	184.8	184.0	184.6	186.9	186.8
283	187	186.75	193.3	184.9	184.8	185.8	184.7	185.6	188.6	186.7
293	186.3	185.89	193.7	185.2	184.8	186.0	184.7	185.8	186.2	185.9
303	184.9	184.3	192.9	184	184.0	185.6	184.0	185.3	185.0	184.4
313	182.7	181.8	191.1	182.1	182.2	184.2	182.3	183.9	184.5	182.1
323	179.6	178.4	188.4	179	179.2	181.8	179.2	181.5	182.1	178.7
333	175.4	173.9	184.6	174.4	174.8	178.2	175.5	177.9	178.2	174.6
343	169.8	168.3	179.9	167.9	168.4	172.8	169.8	172.9	172.3	169.3
353	162.5	161.2	173.2	158.2	159.6	165.2	161.8	166.0	163.2	162.8
363	153	152.5	165.8	145.1	147.0	154.0	156.4	156.4	149.8	154.9
373	140.2	142.07	156.7	123.4	128.2	135.6	132.1	142.1	125.7	145.4
AAD	0.0054	0.0414	0.024	0.020	0.015	0.016	0.0163	0.0193	0.005	

Table 3: Velocity of sound for R 152 a on the boiling curve

T[K]	Exp.	JS	TR	JZG	CYJ	TTL	MMWF
278.5	743	746.1	752.6	749.8	737	699	790
288.5	692	694.9	701.3	701.9	689.9	658.1	744.8
298.5	642	645.2	586.3	600.8	592.8	570.4	650.3
308.5	596	595.4	614	616	606.9	585.7	663.2
318.5	542	544.6	544.8	549	544.6	528.9	600.4
328.5	490	492.8	489.4	491.5	492.6	480.8	546.4
338.5	435	439.6	432.3	430	437.9	429.6	440.2
348.5	382	384.4	373	363.8	379.5	373.8	425.3
358.5	321	326.5	316.1	297.4	320.1	315.9	360.9
368.5	256	263.9	250.3	221	247.9	244.1	287.9
AAD		0.009	0.0217	0.0407	0.0164	0.0376	0.0862